Low-Temperature Properties of $CeRu_4Sn_6$ from NMR and Specific Heat: Heavy Fermions Emerging from a Kondo-Insulating State

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Kondo insulator (KI) materials are Kondo lattice (KL) systems that exhibit semiconducting behavior below a certain temperature T_g at which an energy gap opens at the Fermi level [1,2,3]. Although numerous studies on Kondo insulator compounds have been performed, in which the behavior of the gap under the effect of external parameters like pressure [4] or magnetic field [1] has been investigated, the intrinsic conditions for transforming a KL into a KI are still not clear. A puzzling phenomenon that complicates the concept of a hybridization gap [2] is the fact that an electrically insulating ground state in the limit $T \rightarrow 0$ is rather uncommon. In this group of KI systems a small residual carrier concentration and hence, a finite electrical conductivity for $T \rightarrow 0$ appears to be generic, as indeed observed at low temperature.

The general physical properties of $CeRu_4Sn_6$ are attributed to the formation of the Kondo insulating state in this compound [5,6]. The residual carrier levels in $CeRu_4Sn_6$ are magnetic in origin and yet muon spin relaxation (µSR) experiments [7], a probe which is exceedingly sensitive to magnetically cooperative phenomena, has proven the ground state of $CeRu_4Sn_6$ to be free from long-range or even short-range ordering. $CeRu_4Sn_6$ belongs to a family of rare-earth R-Ru_4Sn_6 ternary stannides [8,9]. Along with U_2Ru_2Sn, it is a rare example of a KI with tetragonal crystal structure. Evidence of the onset of an energy gap at about 30 K has been found in the temperature dependence of several quantities such as the electrical resistivity (ρ) and thermal conductivity (κ) [5,6], the spin-lattice-relaxation rate $^{119}(1/T_1)$ in NMR experiments [10] and the thermopower (S) [5].

^{119}Sn spin lattice relaxation and specific heat

In an effort to investigate the low-energy excitations in $CeRu_4Sn_6$, we have chosen ^{119}Sn-NMR as a microscopic probe, together with the bulk measurement of the specific heat. Both methods are directly linked to the density of states, *N(E)*.

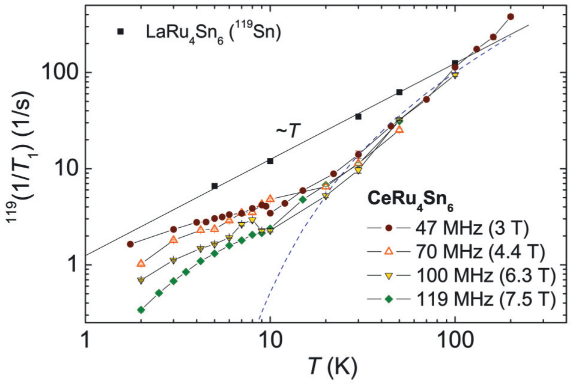

Fig. 1: Temperature evolution of the spin-lattice-relaxation rate $^{119}(1/T_1)$ of $CeRu_4Sn_6$ in a double-logarithmic plot. (Dashed line: $^{119}(1/T_1) \propto T \exp(-\Delta E_g/k_BT)$ with ΔE_g = 30 K). For comparison, the $^{119}(1/T_1) \propto T$ dependence of the non-magnetic homologue $LaRu_4Sn_6$, is also shown [16].

$^{119}(1/T_1)$ vs T of $CeRu_4Sn_6$ deviates significantly from the linear Korringa type of behavior as observed in the reference compound $LaRu_4Sn_6$ (see Fig. 1). This deviation is characteristic of systems exhibiting the opening of an energy gap at the Fermi level upon lowering the temperature. This phenomenon is evident in many other KIs as well, such as U_2Ru_2Sn [11], CeNiSn [12], $Ce_3Bi_4Pt_3$ [13], $FeSb_2$ [14] and SmB_6 [15]. For non-cubic systems like CeNiSn and U_2Ru_2Sn, a T^3-power law was found in $^{119}(1/T_1)$ vs T over two decades in temperature, followed by a residual Korringa-like linear-in-T dependence. A consistent description in the entire temperature range was possible by applying the so-called V-shaped gap model for *N(E)* [12]. For $CeRu_4Sn_6$, however, a T^3-power law behavior was not observed. Furthermore, an unusual and strong field dependence of $^{119}(1/T_1)$ could be found at low temperatures. We attribute this to strongly correlated residual "in-gap" states emerging from the Kondo-insulating state [16].

To study the effect of the residual "in-gap"- states and their possible field and temperature dependencies, a detailed low-temperature specific heat study was performed. In zero field, a logarithmic increase

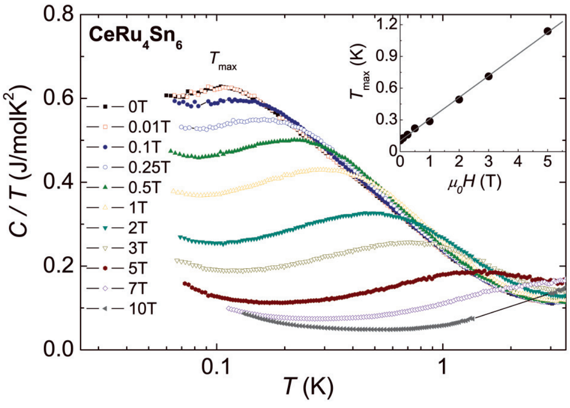

Fig. 2: Sequence of isofield plots of the low-temperature specific heat of $CeRu_4Sn_6$. Inset: Field variation of the temperature T_{max} at which a maximum is achieved in $C(T,H)$, after subtraction of the nuclear contributions [16].

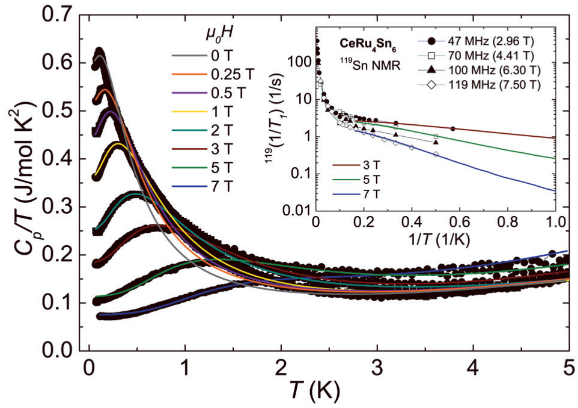

Fig. 3: Temperature dependence of the specific heat (main panel) and spin-lattice-relaxation rate (inset), together with calculations for each applied field, according to the $N(E)$ model presented in Fig 4. [16].

of the Sommerfeld coefficient $\gamma = C/T$ versus T was observed (see Fig. 2). A similar non-Fermi-liquid behavior is often seen in strongly correlated systems (for instance in proximity to a continuous quantum phase transition) and has been the subject of considerable interest.

In the whole temperature range, no sign of a phase transition could be found, in good agreement with µSR measurements [7]. At the lowest temperatures a weak increase of C/T with decreasing T is attributed to a nuclear Schottky contribution. At temperatures sufficiently high compared to the nuclear splitting energy, the high-temperature tail of a Schottky excitation is expected which should follow $C \propto 1/T^2$. In the present case, such a contribution likely arises from ^{119}Sn and ^{117}Sn isotopes as well as from ^{99}Ru and ^{101}Ru isotopes. Furthermore, the nuclear Schottky contribution commonly scales with the magnetic field as H^2, as observed in our case. This low-temperature nuclear contribution is subtracted and is of no further consequence to the interpretation of our data.

$^{119}(1/T_1)(T,H)$ and $\gamma(T,H)$ from a modified $N(E)$-model: comparison with experiment

We have modified the above mentioned V-shaped density-of-states model by replacing the constant and field-independent number of residual states by two field-dependent narrow peaks which are centered around E_F and separated by ΔE_{g2}. Furthermore, the V-shape of the large gap was modified to a conventional U-shape (rectangular), with a temperature and (nearly) field-independent value of $\Delta E_{g1} = 30$ K (see [16] for details on the calculations performed and parameters used). With such an extended model we succeeded in a consistent description of both, the spin lattice relaxation rate $^{119}(1/T_1)$ and the specific heat C/T over the entire temperature range (see Fig. 3).

The density-of-states model used in our calculations for different fields is presented in Figure 4. The behavior is reminiscent of the $S = 1/2$ Kondo-impurity model [17].

The shallow peak in the zero field data of $C(T)/T$ at $T = 0.1$ K or, equivalently, the resulting shallow pseudogap in the zero-field $N(E)$-dependence of the Fermi level suggest some static internal field to develop at the Ce sites possibly due to spin-glass freezing. However, such fields have not been observed at the muon-stopping sites in µSR experiments down to $T = 0.05$ K [7]. We can now use this density of states to calculate all the observables. The point N_0 at which $N(E)$ crosses the Fermi level provides the Sommerfeld coefficient γ at $T = 0$. Plotting N_0 as a function of $log(\mu_0 H)$ indicates an exponential decreases of γ with increasing magnetic field (see inset (a) of Fig. 4). To compare the values of $\Delta E_{g2}/k_B T$ obtained from NMR measurements with those obtained by fitting the specific-heat data, we plotted both datasets in the same diagram, *cf.* inset (b) of the same Figure. Similar values and a comparable field dependence are found for the two different approaches. Assuming a Zeeman energy of $\Delta E = -\mu B$, the magnetic moment of these states can be estimated from the slope of the dashed line in the inset (b) to Figure 4. The fit gives a value close to 1 μ_B. To estimate the number of states per formula unit, $N(E)$ can be inte-

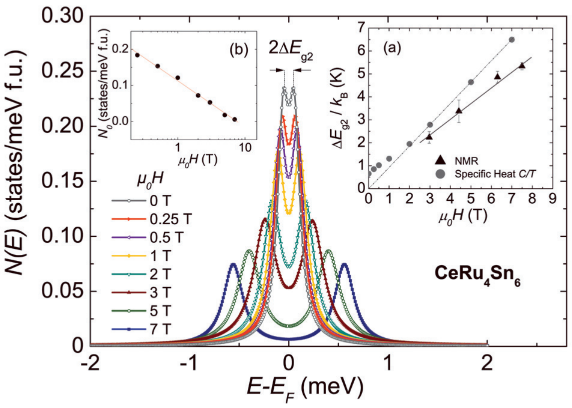

Fig. 4: Density of states projection against energy relative to the Fermi energy E_F. The field-derived $T \to 0$ density of states is depicted in inset (b) whereas inset (a) plots the field evolution of the inner gap ΔE_{g2} responsible for the low-energy properties. Lines are guides to the eye [16].

grated. At zero field and $T = 2$ K, one obtains 0.039 states per formula unit. This value is in good accordance with the one obtained from Hall-coefficient data, which gave 0.03 carriers per formula unit [5].

Conclusion

We applied our *N(E)*-model to the specific-heat and the NMR relaxation data of $CeRu_4Sn_6$ with reasonable success. Specifically the field dependence of these properties was found to be fully reconcilable in the framework of this model. The temperature evolution of electronic and thermodynamic properties is driven by two nested energy gaps, ΔE_{g1} = 30 K and $\Delta E_{g2} \approx 0.65$ K (at zero field), centered around the Fermi energy. The gap is not completely formed, and a residual number of heavy-mass charge carriers within the smaller of the two gaps gives rise to a finite electrical conductivity even at the lowest temperatures. These states are correlated, presumably due to the Kondo-screening effect, as inferred from strong temperature dependence of the Sommerfeld-coefficient of the 4f increment to the specific heat and the magnetic susceptibility, both of which are found to saturate at large values as $T \to 0$. The scope of the physics put forward for $CeRu_4Sn_6$ in this work provides a platform from which the predicted nodal Kondo insulating state as a new type of semimetal and its formation under favorable conditions can be tested. The axial symmetry of nodes at which the density of states vanishes sets the stage for anisotropic electronic conduction. Anisotropic magnetic properties in $CeRu_4Sn_6$ single crystals have recently been reported [18], and our studies are also extended to single crystal material. As a first step we investigate the anisotropic electrodynamic response of single crystals [18] along the *a* and *c* axes in the frequency range from 5 meV to 0.85 eV at various temperatures down to 5 K. For both polarizations the optical conductivity is strongly frequency and temperature dependent over a broad energy range. We observe a strong and anisotropic depletion of the spectral weight at low temperatures up to 0.1 eV, which is more pronounced along the *a* axis. Finally, our results may well reflect the interplay between RKKY and Kondo interaction in the limit of very low carrier concentration as discussed by Coqblin et al. [19].

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